THE U.S. OFFICE OF NAVAL RESEARCH

Grant N00014-94-1-0053

October 1, 1993 to September 30, 1996

Final Technical Report

INVESTIGATION OF MICROSCOPIC MECHANISMS OF FAILURE OF ELECTRONIC SMART MATERIALS/SYSTEMS

PRINCIPAL INVESTIGATOR: Qing Jiang

Department of Engineering Mechanics

Center for Materials Research and Analysis

University of Nebraska-Lincoln

Lincoln, Nebraska 68588-0347

Tel. 402-472-8858, Fax. 402-472-9282

19970421 052

DISTRIBUTION STATEMENT A

Approved for patitive relocates

Distribution Unlimited

DTIC QUALITY INSPECTED 3

UNIVERSITY OF NEBRASKA-LINCOLN

FINAL TECHNICAL REPORT

ONR Grant N00014-94-1-0053

Principal Investigator: Qing Jiang

1. Introduction

The development of ferroelectric ceramics is driven by the needs of functional ceramics for various applications, such as sensors, transducers and actuators, and these functional ceramics account for more than 60% of the total high technology ceramics market worldwide. Owing to their strong electromechanical coupling effect and the prompt response to applied electric fields, ferroelectric ceramics have been increasingly used in designing smart actuators for active control applications, such as large flexible space trusses (Crawley and de Luis 1987), fixed wing and helicopter rotary blades (Sprangler and Hall 1990, Samak and Chopra 1993 & 1994, Giurgiutiu, Chaudhry and Rogers 1994) and automotive suspensions (Thirupathi and Naganathan 1992). The most commonly used ferroelectic ceramics for transducer and actuator applications are the oxides of lead zirconium titanium, $Pb(Zr_x, Ti_{1-x})O_3$, also known as PZT ceramics, because of their strong electromechanical coupling effect and relative low cost for massive production. The actuation force, or the actuation displacement equivalently, is determined by a material parameter, named the remnant polarization which is related to the electro-mechanical coupling effect of polycrystalline ferroelectric ceramics. The remnant polarization of PZT ceramics deteriorates after many cycles of applied electric field (Jiang, Cao and Cross 1994) and, consequently, the actuation force or the actuation displacement diminishes. This phenomenon, being referred to as *electric fatigue*, severely restricts the operational life of these actuators.

This project focused on the microscopic mechanisms of electric fatigue of ferroelectric ceramics. The present report summarizes this investigation and the major results along with the detailed analysis have been documented in a number of publications which are listed in the references of this report.

2. The Microstructure of PZT and Its Evolution during Processing

PZT is a generic term for a family of solid solutions $Pb[Zr_xTi_{1-x}]O_3$ of lead titanate and lead zirconate, being a binary system (Deri 1969, Jaffe, Cook and Jaffe 1971, Saito 1988, Xu 1991, Cross 1992). At room temperature, PZT has either a tetragonal structure (on the Ti-rich side) or a rhombohedral structure (on the Zr-rich side), depending on the molar ratio, except on the extreme Zrrich side where the solid solution has an orthorhombic structure, exhibiting no observable electromechanical coupling effect. The commonly used PZT ceramics have a tetragonal structure with a composition near the morphotropic phase boundary because of the strong electromechanical coupling effect in this region. The tetragonal structure is polar in the sense that the centers of positive and negative charges for each lattice unit are spatially separated, forming a dipole of electric charges. Consequently, these crystals are polarized even in the absence of applied electric fields, a phenomenon referred to as spontaneous polarization. In a stable configuration, a crystallite of PZT ceramic is divided into a number of macroscopic regions in which the polar directions differ from each other by either 180° or approximately 90°†. These regions are called the ferroelectric domains and correspondingly, the interfaces are referred to as the 180° or 90° domain

[†] The deviation from 90° is determined by the lattice parameters.

walls, respectively. For a polycrystalline ceramic, the macroscopic polarization is measured by the average of polarization across the ceramic, called remnant polarization.

Application of an electric field can switch the polar direction of a ferroelectric domain by either 180° or 90°, called 180° or 90° domain switching, respectively, and the resulting polar direction is closer to the direction of the applied electric field. 90° domain switching is accompanied by lattice distortions and the resulting deformation is referred to as *spontaneous deformation*. In response to application of an electric field, domain switching takes place, and the associated lattice distortion leads to a macroscopic deformation or a force when the ceramic is clamped. This provides the mechanism for electrically-controlled actuation.

The commercial PZT ceramics are produced as polycrystalline solid solutions through the conventional steps of sintering of fine powders of oxide metals, PbO, ZrO_2 and TiO_2 . The solid solutions resulting from sintering do not exhibit observable electromechanical coupling effect because the ferroelectric domains are so formed that the average polarization of each grain is approximately zero. After sintering, a strong DC electric field is applied to the semi-finished products, to force domain reorient towards the direction of the applied electric field, resulting in a remnant polarization. This is the so-called *poling process*.

3. The Stress and Electric Field Concentrations

With a premilinary analysis within the framework of lattice mechanics, the PI (Jiang 1994a) shows that interaction of ferroelectric domains with grain boundries can result in stress concentrations at the intersections of domain walls with grain boundaries due to deformations associated with domain formation and switching. In order to carry out detailed analysis on the stress concentrations of this type, the PI have developed a mathematical model (Jiang 1994b& c) characterizing macroscopic behavior of ferroelectric ceramics under electrical and/or mechanical loading programs, taking into account the microstructural transformations associated with formation of ferroelectric domains. A number of predictions of this model, such as the electrically-controlled stress relaxation (Jiang 1994d & 1995a), the ferroelectric domain morphology (Rosakis and Jiang 1995) and the effect of stresses upon the microstructural transformation (Jiang 1995b), are in qualitative agreement with experimental observations. The PI and his associates then applied this model to investigate the stress concentration at intersections of ferroelectric domain walls with grain boundaries. Their analysis (Jiang and Zhang 1994) shows that the stress concentration is always accompanied by electric field concentration because of the strong electromechanical coupling effect and it corresponds to the so-called power-law singularity, i.e., the magnitudes of stress and electric fields near such a intersection are proportional to r^{λ} , with r being the distance to the intersection and λ being such a constant that $-1 < \lambda < 0$. The order parameter λ , which measures the severity of the concentration, depends upon the crystal polar direction and the orientation of the domain walls with respect to the grain boundary which they intersect. Under a cyclic loading program, microcracks can be initiated at these intersections when the stress exceeds the material strength locally. To understand the effects of ferroelectric domains on crack growth, Zhang and Jiang (1995a & b) have also investigated interactions of ferroelectric domains with pre-existing microcracks, both intergranular and transgranular, and their analysis shows that the interaction makes the stress concentrations near the crack tips substantially stronger than the so-called \sqrt{r} -singularity which appears in the absence of the domain wall interaction. This suggests that the interaction promotes the growth of microcracks. The predicted stress distribution indicates that microcracks can appear as clusters near tips of pre-existing cracks, a phenomenon recently observed by Hill, White, Hwang and Lloyd (1996). Wang and Jiang (1995) have investigated the mechanical analogy of the domain wall interaction, i.e., the interaction of mechanical twinning with cracks in non-ferroelectric materials, such as α -iron and copper zirconate. Their analysis shows that magnification of stress concentrations at crack tips due to twinning is a general phenomenon characteristic of polycrystalline solids.

4. Domain Switch in Polycrystalline Ferroelectric Ceramics

The PZT ceramics are produced as polycrystalline solid solutions through hot-forging, hot-pressing and sintering of fine powders of lead titanate and lead zirconate. The resulting solid solutions do not exhibit observable electromechanical coupling effect because of the random orientation of grains. The coupling effect appears only after the grains are reoriented by application of a strong DC field. This reorientation process is the so-called *poling process*. Ferroelectric domains are initially formed when the ceramics are cooled from the high processing temperature and they are altered during the subsequent poling process. Application of an electric field can switch the polar direction of a ferroelectric domain, called the *domain switch* or *polarization switch*. Domain switch results in a macroscopic displacement or force which provides the actuation mechanism. Stress concentrations are magnified during both the cooling process and the actuator operational process because of domain switch. The PI and his associates have modeled the domain switch in polycrystalline ferroelectric ceramics for both the poling process, where the ceramics gain the electromechanical coupling ef-

fect (Huo and Jiang 1995b, Huo and Jiang 1996), and the actuator operational process, where the macroscopic polar direction switches periodically under cyclic electric-loading programs (Wang, Gong and Jiang 1995).

5. Dynamic Effect of Polarization Switch

Considering that ferroelectric actuators are operated at high frequency for some applications, the PI and his associates have investigated vibration of a ferroelectric plate under excitation of an AC field, taking into account the inertia effect. To investigate the effect of domain switch, they have determined the electrically-induced internal stresses for both cases: with and without domain switch. Their analysis (Huo and Jiang 1995a) concludes that domain switch results in stresses of frequency close to resonance, independent of the frequency of the applied electric field, and of a substantially larger amplitude than stresses occurring in the absence of domain switch. This explains the phenomenon recently observed by Jiang, Cao and Cross (1994) that the frequency of applied AC field has no significant effect on fatigue life.

6. The Mechanisms of Electric Fatigue

It is known that fatigued ferroelectric materials often contain many microcracks (Pohanka, Rice and Walker 1976, Pohanka, Freiman and Bender 1978, White, Hill, Hwang and Freiman 1995). Jiang and Cross (1993) recently observed that the remnant polarization in a fatigued PZT specimen partially recovered after being heated at $300^{\circ}C$ for two hours. This observation suggests that microcracking may not the sole cause of electric fatigue because the temperature is too low for healing microcracks in the PZT specimen. The other cause may be the so-called domain pinning which refers to the notion that some domains become no longer switchable. This notion was first introduced by Merz and

Anderson (1955) to explain, qualitatively, the thickness dependence of dynamic properties of ferroelectric specimen but it has never been verified. Wang, Gong and Jiang (1995) have recently developed a computational model to simulate the effect of domain pinning on remnant polarization and their results confirm that domain pinning can indeed cause fatigue behavior of the type observed by Jiang, Cao and Cross (1994).

7. Effect of Microcracks upon Electric Fatigue

To understand the role of microcracking in electric fatigue, the PI and his associates have carried out a numerical analysis, to simulate the effect of microcracks on the macroscopic properties. Kim and Jiang (1995) have developed a finite element algorithm based on a fully-coupled theory of ferroelectric polycrystalline ceramics (Jiang 1994b& c, Jiang and Zhang 1994). Considering that the size of these microcracks is comparable to the grain size and that they usually grow along grain boundaries, Kim and Jiang (1995) have designed such an algorithm that is particularly suitable for modeling the effects of grain boundaries and intergranular cracking. Their numerical results indicate that formation and subsequent growth of microcracks lead to redistribution of the electric field within the actuators, resulting in migration of electric charges. As a result of accumulation of electric charges on the crack faces, some domains or grains near microcracks become pinned. The remnant polarization, being a macroscopic measure of spontaneous polarization of the actuator, deteriorates as pinned domains are developed and correspondingly, the actuation displacement diminishes.

References

Crawley, E.F. and J. de Luis, 1987, Use of piezoelectric actuators as elements of intelligent structures, *AIAA Journal*, Vol. 5, pp. 1373-1385.

Giurgiutiu, V., Z. Chaudhry and C.A. Rogers, 1994, Engineering feasibility of induced strain actuators for rotor blade active vibration control, *Proceedings* of the 1994 SPIE Smart Structures and Materials Conference, Orlando, Florida, pp. 107-122.

Hill, M.D., G.S. White, C.S. Hwang and I.K. Lloyd, 1996, Cyclic damage in lead zirconate titanate, *Journal of the American Ceramic Society*, vol. 79, pp. 1915-1920.

Huo, Y.Z. and Q. Jiang, 1995a, Effect of polarization switch upon stress in thickness vibration of a ferroelectric plate, **Technical Report No. 2**, ONR Grant N00014-94-1-0053, also *Journal of the American Ceramic Society*, vol. 79, pp. 651-654.

Huo, Y.Z. and Q. Jiang, 1995b, A constitutive model for domain switch in polycrystalline ferroelectric ceramics, **Technical Report No. 5**, ONR Grant N00014-94-1-0053, also *Smart Materials & Structures*, in press.

Huo, Y.Z. and Q. Jiang, 1996, Modeling of domain switching in ferroelectric ceramics: An example, *International Journal of Solids and Structures*, in press.

Jiang, Q., 1994a, On the mechanisms of microcracking in ferroelectric ceramics, Proceedings of the Second International Conference on Intelligent Materials, pp. 677-685, Technomic Publishing, Lancaster.

Jiang, Q., 1994b, On modeling of phase transformations in ferroelectric materials, *Acta Mechanica*, Vol. 102, pp. 149-165.

Jiang, Q., 1994c, On the driving traction acting on a surface of discontinuity within a continuum in the presence of electromagnetic fields, **Technical Report**No. 1, ONR Grant N00014-94-1-0053, also *Journal of Elasticity*, Vol. 34, pp. 1-21.

Jiang, Q., 1994d, Modeling of stress relaxation associated with an electrically induced phase transformation, **Technical Report No. 2**, ONR Grant N00014-94-1-0053, also *Journal of Applied Mathematics and Physics*, Vol. 45, pp. 899-913.

Jiang, Q., 1995a, On the electro-mechanical response of electrically active materials, *Journal of Intelligent Material Systems and Structures*, Vol. 6, pp. 181-191.

Jiang, Q., 1995b, A lattice-based model for the cubic-tetragonal transformation in ferroelectric crystals, **Applied Mechanics in the Americas**, Vol. 1, pp. 455-460, American Academy of Mechanics.

Jiang, Q., W. Cao and L.E. Cross, 1994, Electric fatigue in lead zirconate titanate ceramics, *Journal of the American Ceramic Society*, Vol. 77, pp. 211-215.

Jiang, Q. and L.E. Cross, 1993, Effect of porosity on electric fatigue behavior in PLZT AND PZT ceramics, *Journal of Material Science*, Vol. 28, pp. 4536-4543.

Jiang, Q. and Y. Zhang, 1994, Microcracking in ferroelectric ceramics: Part 1. Ferroelectric twinning, **Technical Report No. 5**, ONR Grant N00014-94-1-0053, also **Active Materials and Smart Structures**, SPIE Publications, Vol. 2427, pp. 310-325.

Kim, S.J. and Q. Jiang, 1995, Modeling of microcracking in polycrystalline ferroelectric ceramics, **Technical Report No. 6**, ONR Grant N00014-94-1-0053, also *Smart Materials & Structures*, Vol. 5, pp. 321-326.

Merz, W.J. and J.R. Anderson, 1955, Bell Laboratory Record, Vol. 33, p. 335.

Pohanka, R.C., S.W. Freiman and B.A. Bender, 1978, Effect of the phase transformation on the fracture behavior of $BaTiO_3$, Journal of the American Ceramic Society, Vol. 61, pp. 72-75.

Pohanka, R.C., R.W. Rice and B.E. Walker, 1976, Effect of internal stress on the strength of $BaTiO_3$, Journal of the American Ceramic Society, Vol. 59, pp. 71-74.

Rosakis, P. and Q. Jiang, 1994, On the morphology of ferroelectric domains, **Technical Report No. 3**, ONR Grant N00014-94-1-0053, also *International Journal of Engineering Science*, Vol. 33, pp. 1-12.

Samak, D.K. and I. Chopra, 1993, A feasibility study to build a smart rotor: trailing edge flap actuation, *Proceedings of the 1993 North American Conference on Smart Structures and Materials*.

Samak, D.K. and I. Chopra, 1994, Design of high force, high displacement actuators for helicopter rotors, *Proceedings of the 1994 North American Conference on Smart Structures and Materials*.

Sprangler, R.L. and S.R. Hall, 1990, Piezoelectric actuators for helicopter rotor control, *Precedings of AIAA/ASME/ASCE/AHS/ASC 31st Structures, Structural Dynamics and Materials Conference*, pp. 1589-1599.

Thirupathi, S.R. and G.G. Naganathan, 1992, Use of piezoelectric actuation for automotive active suspension mechanisms: A feasibility study, *Proceedings of the 22nd Biennial Mechanism Conference*, pp. 233-241.

Wang, Y.Q. and Q. Jiang, 1995, Twinning-induced stress concentrations in polycrystalline solids, **Technical Report No. 3**, ONR Grant N00014-94-1-0053, also *Mathematics and Mechanics of Solids*, Vol. 1, pp. 95-110.

Wang, Y.Q., S.X. Gong, and Q. Jiang, 1995, Effect of domain pinning on electric fatigue in ferroelectric ceramics, **Technical Report No. 4**, ONR Grant N00014-94-1-0053, also *Ferroelectrics*, Vol. 182, pp. 61-68.

Zhang, Y. and Q. Jiang, 1995a, Microcracking in ferroelectric ceramics: Part 2. Intergranular and transgranular cracking, **Technical Report No. 1**, ONR Grant N00014-94-1-0053, also **Mathematics and Control in Smart Structures** SPIE Publications, Vol. 2442, pp. 11-22.

Zhang, Y. and Q. Jiang, 1995b, Twinning-induced stress and electric field concentrations in ferroelectric ceramics, *Journal of the American Ceramic Society*, Vol. 78, pp. 3290-3296.